

DECLARATION OF KOUICHI TAKEMOTO  
UNDER 37 CFR §1.132

I, Kouichi Takemoto, having been wanted in accordance with Section 1001 of Title 18 of the United States Code, declare that: I have 12 years of experience in the research and development of electronic materials in Nippon Mining & Metals Co., Ltd.

I am familiar with the properties of metals commonly used to manufacture semiconductor devices. In particular, I am skilled in the art of producing high purity copper sulfate for using copper electroplating upon manufacturing semiconductor devices.

I am one of the inventors of the present invention. I will explain the differences in the manufacturing method and characters of the copper sulfate between the present invention and prior references: JP 05-262523, JP47-040634 and U.S. Patent No. 5,059,403.

Comparison with D1 (JP 05-262523)

The object to be achieved by the invention of Document 1 is as follows: "To provide a method of producing, at low cost and in a short time, a copper sulfate solution directly from metallic copper powder without going through copper oxide powder (excerpt from original text)" (paragraph 0004). In order to achieve the foregoing object, Document 1 is characterized in performing the following processes: "While introducing large amounts of fine air bubbles in the suspension of metallic copper powder, sulfuric acid is added while maintaining the solution between 65°C and 85°C, and metallic copper is oxidized and melted (excerpt from original text)" (paragraph 0005). In addition, the Example (Experiment No. 1) of Document 1 describes as follows: "920L of industrial water was placed in a melting unit comprising a bath with a jacket having a volume of 1.5m<sup>3</sup> and an air-type agitator, 125Kg of metallic copper powder having a grain size of 40 mesh to 60 mesh was placed therein and suspended, rotation of the agitator was set to 125rpm, and 225Kg of 98% concentrated sulfuric acid was added over a period of approximately one hour. The liquid temperature was maintained at 70°C during this period. Agitation was continued for an additional two hours. The inversion rate to copper sulfate was 99.2% (excerpt from original text)." Moreover, in Experiment No. 2 it is described as follows: "Metallic copper powder was placed in a solution containing 18g/L of arsenic, 30g/L of copper, and 90g/L of sulfuric acid and suspended (excerpt from original text)." Meanwhile, although the purity level of the

copper sulfate solution is affected by the purity level of the raw material copper powder in Document 1, Document 1 does not in any way describe the purity of copper sulfate.

In light of the above, since the invention of Document 1 performs "air blowing" in order to oxidize and melt the metallic copper, air dust (impurities) gets mixed into the suspension. Moreover, in Experiment No. 1 of the Examples, industrial water is used as the suspension, and it is evident that no consideration is given to the purity. Moreover, since Experiment No. 2 uses arsenic, the inclusion of arsenic is unavoidable. From the foregoing facts, not only does Document 1 fail to clearly specify copper sulfate having a purity of 99.99% or higher, it is obvious that it is not possible to produce copper sulfate having a purity of 99.99% or higher from Document 1. Thus, the novelty (anticipation) of the present invention cannot be denied based on Document 1.

#### Comparison with D2 (JP 47-040634)

Document 2 describes as follows: "The present inventors focused attention on the fact that it is possible to sulfate the copper content in the copper sulfate ore nearly completely by fluidizing and roasting the copper sulfide ore, and thereby conducted research for a method of economically producing copper sulfate by extracting copper sulfate from the copper sulfide ore that was subject to sulfation and roasting (excerpt from original text)." Moreover, as the production method of copper sulfate, Document 2 describes as follows: "A method of producing copper sulfate, comprising: (1) a first step of sulfating and roasting copper sulfide ore, (2) a second step of extracting copper sulfate under calcination as a copper sulfate tetraamine solution with a diluted aqueous ammonia and separating it from residue, (3) a third step of increasing the ammonia concentration in the obtained extraction liquid to precipitate crystals of copper sulfate tetraamine and separating them from the mother liquid, and (4) a fourth step of heating the obtained crystals to 400 to 800°C to obtain anhydrous copper sulfate (excerpt from original text)." In addition, the Example of Document 2 describes as follows: "100g of copper concentrate (Cu 18.84%, Fe 30.47%, S 29.68%, SiO<sub>2</sub> 8.20%, CaO 4.46%, Zn 067%, Pb 0.24%) was used, ...sulfation and roasting were performed to obtain 117g of sintered mine (cited from original text)" and "The anhydrous copper sulfate contains 39.5% of copper, and corresponds to a purity of 99.3% as the copper sulfate (excerpt from original text)."

As described above, with the invention pertaining to Document 2, metallic impurities such as Fe, Zn and Pb are contained in the copper concentrate as the raw material. Further, the purity that is achievable in the invention of Document 2 is 99.3%, and it is evident that impurities are contained in the refined copper sulfate at a ratio of 0.7%. Thus, Document 2 has no technical justification of being able to achieve 99.99%. Moreover, the usage of copper sulfate that is anticipated in Document 2 is as follows: "agricultural chemical (Bordeaux mixture), antiseptic agent (railroad crosstie, utility pole, starch for spinning, fishing net), Bemberg rayon, iodine production, ceramic material, pigment (excerpt from original text)." Accordingly, Document 2 clearly has a different recognition on purity in comparison to the present invention which aims to improve the plating characteristics of a microstructure semiconductor device.

#### Comparison with D3 (US 5,059,403)

Document 3 describes that the lower limit of the copper sulfate purity is 99.8%. Meanwhile, the upper limit of purity is unclear since it is not described anywhere in the specification including the Examples. The problem with Document 3 is that it only discloses the lower limit and fails to disclose the upper limit. Specifically, although it is possible to assume that the lower limit is approximately 99.8% or in the vicinity thereof, so as long as there is no clear verification of being able to improve the purity level to be higher, it is not possible to determine that Document 3 is able to achieve a purity level of 99.99%.

In other words, it would be erroneous to assume that 99.99% is included in the purity range that can be achieved by Document 3 in which the achievable upper limit of purity is unclear. In addition, Document 3 entails factors that deteriorate the purity level as described below.

The purification technology of Document 3 is characterized in collecting copper as copper sulfate from waste liquid with numerous impurities. In addition, since a product obtained by neutralizing the copper sulfate solution with sodium hydroxide is added once again, there is no recognition of eliminating Na as an impurity. In addition, since Document 3 does not perform the treatment of eliminating the early precipitation crystals as with the present invention, in the least, the impurities that should be eliminated by the foregoing

treatment (treatment of eliminating the early precipitation crystals) will remain. Accordingly, it is impossible to assume that Document 3 is able to achieve the same level of purity as the present invention.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like are punishable by fine and imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date: *Jan 17 / 2010*

KOUCHI TAKEMOTO

Signature: *Kouchi Takemoto*